

Upper critical field and de Haas-van Alphen oscillations in KOs_2O_6 measured in a hybrid magnet

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Magnetic torque measurements have been performed on a KOs_2O_6 single crystal in magnetic fields up to 35.3 T and at temperatures down to 0.6 K. The upper critical field is determined to be ~ 30 T. De Haas-van Alphen oscillations are observed. A large mass enhancement of $(1+\lambda) = m^*/m_{\text{band}} = 7.6$ is found. It is suggested that, for the large upper critical field to be reconciled with Pauli paramagnetic limiting, the observed mass enhancement must be of electron-phonon origin, including electron-rattling-mode interactions, for the most part.

KEYWORDS: pyrochlore oxides, KOs_2O_6 , upper critical field, de Haas-van Alphen effect

The β -pyrochlore osmium oxides AOs_2O_6 ($A = \text{K}, \text{Rb}, \text{and Cs}$) exhibit superconductivity with the transition temperatures T_c of 9.6, 6.3, and 3.3 K, respectively.^{1–4} In a recent paper,⁵ Hiroi and coworkers have argued that pairing in these compounds is mediated by a low-energy rattling mode. A^+ ions in AOs_2O_6 are enclosed in much bigger cages formed by OsO_6 octahedra and vibrate with large amplitudes in an anharmonic potential.⁶ Such vibrations are called rattling. This unique variant of electron-phonon superconductivity deserves further detailed studies.

We here report magnetic torque measurements on KOs_2O_6 . We focus on two subjects. The first is the upper critical field B_{c2} . We confirm a large upper critical field at zero temperature $B_{c2}(0) \sim 30$ T as previously reported.^{7,8} For comparison, the paramagnetic critical field B_{po} is 18 T if evaluated as $B_{po} = 1.84T_c$ (B_{po} in Tesla and T_c in Kelvin) as usual. To resolve this apparent contradiction, a previous interpretation invoked noncentrosymmetry of the crystal structure.^{7,9} However, it is now clear that the crystal structure remains centrosymmetric down to low temperatures below T_c .^{10–13} Accordingly, we reexamine if and how the large $B_{c2}(0)$ can be reconciled with Pauli limiting. The other is the de Haas-van Alphen (dHvA) effect. We have observed dHvA oscillations in KOs_2O_6 for the first time. From comparison with band structure calculations, we find an unusually large mass enhancement for the basically electron-phonon superconductor, confirming previous analyses of specific heat data.^{5,14} Since the specific heat was measured only up to 14 T, where T_c is still 5.2 K, the Sommerfeld coefficient γ at low temperatures could not be determined directly but was estimated on the basis of a few assumptions including an unconventional temperature dependence of the lattice contribution. Therefore the present direct confirmation is invaluable.

The single crystal used in the present study was prepared as described in ref. 14, where a residual resistivity ratio of about 300 was reported. Magnetic fields up to 35.3 T were produced by the hybrid magnet installed at

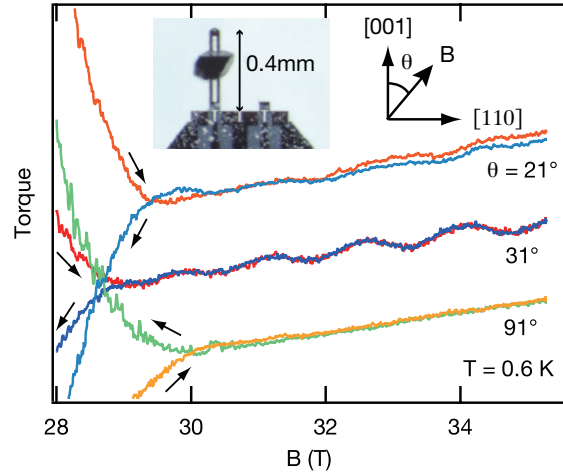


Fig. 1. (Color online) Magnetic torque of KOs_2O_6 at 0.6 K for three different field directions. Both field up and field down sweeps are shown. The left inset shows the single crystal sample mounted on a microcantilever, and the right inset shows the definition of the field angle θ .

the Tsukuba Magnet Laboratory of the NIMS.¹⁵ Low temperatures down to 0.6 K were generated by a ^3He refrigerator. Magnetic torque was detected by using a piezoresistive microcantilever (Fig. 1). The field direction θ is measured from $[001]$ toward $[110]$ (Fig. 1). The band structure calculations were performed within the local density approximation using a full potential LAPW (FLAPW) method.¹⁶ We used the program codes TSPACE¹⁷ and KANSAL-06. The obtained electronic band structure is very similar to that previously obtained for CsOs_2O_6 .¹⁸

Figure 1 shows magnetic torque measured at 0.6 K as a function of field for three field directions, where both field up and field down sweeps were made. The up- and down-field traces diverge below 30 T for $\theta = 21$ and 91° . For $\theta = 31^\circ$ the divergence occurs at a slightly lower field, i.e., 29 T. We identify these fields with $B_{c2}(0)$ and conclude

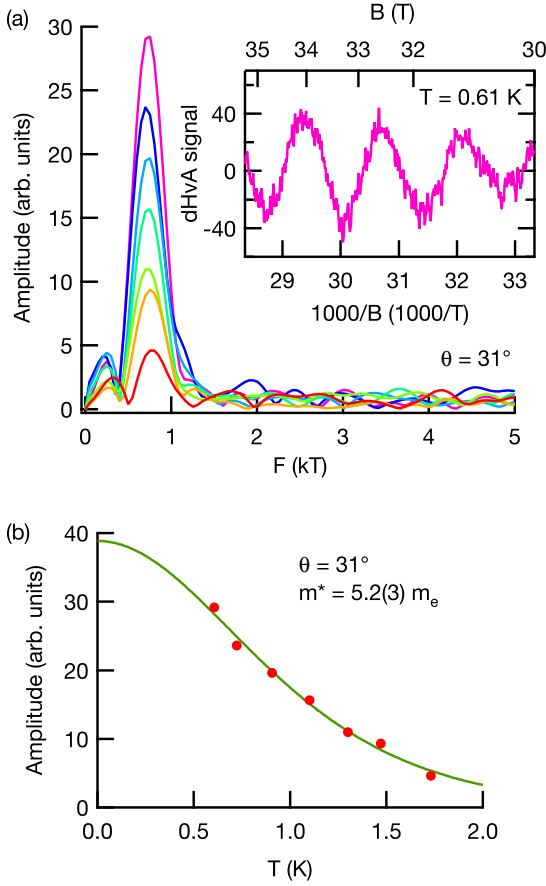


Fig. 2. (Color online) (a) The inset shows the dHvA oscillatory torque at $T = 0.61$ K and $\theta = 31^\circ$ obtained after smoothly-varying background was subtracted from the data in Fig. 1. The up- and down-sweep data were averaged. The main part shows Fourier transforms of oscillations (in $1/B$) at $\theta = 31^\circ$ for various temperatures. $T = 0.61, 0.72, 0.91, 1.1, 1.3, 1.5$, and 1.7 K with decreasing amplitude. (b) Temperature dependence of the oscillation amplitude. The solid curve is a fit to the Lifshitz-Kosevich formula, from which we deduce that $m^* = 5.2(3) m_e$.

that $B_{c2}(0) \sim 30$ T, assuming $B_{c2}(0) \approx B_{c2}(0.6\text{K})$.¹⁹⁾ This estimation is close to ~ 33 and 30.6 T reported in refs. 7 and 8, respectively. The smaller field value observed for $\theta = 31^\circ$ may indicate the anisotropy of B_{c2} , which can exist even in a cubic material if the Fermi surface is nonspherical.²⁰⁾

dHvA oscillations are already evident for $\theta = 21$ and 31° above B_{c2} even before smoothly-varying background is subtracted. The dHvA oscillatory torque at $\theta = 31^\circ$ obtained after the background subtraction and the corresponding Fourier transform are shown Fig. 2(a). A single peak appears at a dHvA frequency $F = 0.73$ kT in the transform. The angle dependence of F is shown in Fig. 3. $F = 0.54(2)$ kT for $B \parallel [001]$. The observed frequency branch is in an excellent agreement with the calculated β branch, which was named after the corresponding branch in CsOs_2O_6 .¹⁸⁾ The β orbit is on the hole surface as shown in Fig. 4.

The temperature dependence of the dHvA oscillation amplitude measured at $\theta = 31^\circ$ is shown in Fig. 2. The effective mass m^* is estimated from a fit to the Lifshitz-

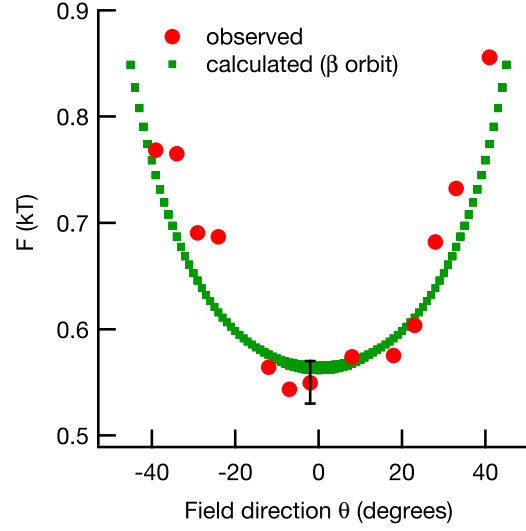


Fig. 3. (Color online) Angle dependence of the observed frequency compared to the calculated β frequency.

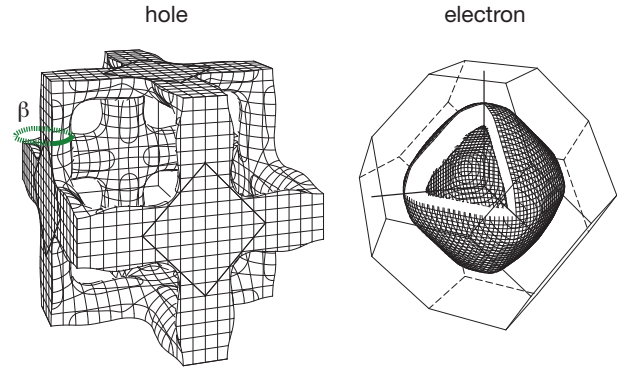


Fig. 4. (Color online) Calculated Fermi surface of KOs_2O_6 . The experimentally observed β orbit is on the hole sheet.

Kosevich formula²¹⁾ to be $5.2(3) m_e$, m_e being the free electron mass. According to our band structure calculations, the band mass m_{band} is $0.68m_e$. Thus the mass enhancement $(1+\lambda)$ is estimated to be $(1+\lambda) = m^*/m_{band} = 7.6$, which is very close to the value of 7.3 estimated from the specific heat data.⁵⁾ In the case of CsOs_2O_6 , the mass enhancement is 4.2 for the β orbit.¹⁸⁾ Thus the mass enhancement for the β orbit is increased by 81% from CsOs_2O_6 to KOs_2O_6 . This is also consistent with a 94% increase in the specific heat mass enhancement $[(1+\lambda) = 3.76$ from the specific heat in CsOs_2O_6].⁵⁾

Three points should be noted here. Firstly, the observed mass enhancement of 7.6 is unusually large when compared to values found in other superconductors with relatively high T_c 's. dHvA measurements on $\text{LuNi}_2\text{B}_2\text{C}$ ($T_c = 16$ K) and MgB_2 ($T_c = 38$ K) have observed, respectively, the mass enhancements $(1+\lambda)$ of 1.3 – 3.7 ²²⁾ (or 1.16 – 3.02 ²³⁾) and those of 1.31 – 2.2 ,²⁴⁾ depending on orbits. The mass enhancements of 2 – 3 have been found for the A15 compounds Nb_3Sn ($T_c = 18$ K) and V_3Si ($T_c = 17$ K) and the Chevrel compounds SnMo_6S_8 and

PbMo_6S_8 ($T_c = 12$ K for both) from analyses of specific heat.^{25,26)}

Secondly, the values of m^* and γ were estimated at very different magnetic fields: m^* 's were estimated at 9.3 and 32 T for CsOs_2O_6 and KOs_2O_6 , respectively, while γ 's were estimated for zero field. The observed consistency among these data indicates that the mass enhancement is insensitive to the magnetic field.

Thirdly, the observed consistency also suggests that the mass enhancement is fairly homogeneous over the Fermi surface. At least, the mass enhancement does not vary as it does in $\text{LuNi}_2\text{B}_2\text{C}$, where it varies from 1.3 to 3.7²²⁾ (or 1.16 to 3.02²³⁾) depending on orbits.

No other dHvA frequency than β has been observed in the present study. This can basically be explained by results of the band structure calculations: Namely, band masses associated with other possible dHvA frequencies are heavier than the mass of β . Because of relatively high measurement temperatures ($T \geq 0.6$ K), dHvA oscillations are rapidly suppressed as effective masses increase. In addition, since amplitude of dHvA torque oscillation is proportional to $\ln F/d\theta$, it is difficult to detect dHvA frequency branches with little angular dependence, like the γ and δ branches in CsOs_2O_6 ,¹⁸⁾ in torque measurements.

Having determined the mass enhancement in KOs_2O_6 , we now return to the large B_{c2} . As one goes from CsOs_2O_6 to KOs_2O_6 , $B_{c2}(0)$ increases from 1.4¹⁸⁾ to ~ 30 T. Since T_c increases only by a factor of three, this factor-of-21 increase may appear surprising at first glance. However, this increase is consistent with an expected increase in the orbital critical field $B_{c2}^*(0)$. In the clean limit of an isotropic single-band model, $B_{c2}(0)^* \propto \gamma^2 T_c^2 = \gamma_{band}^2 (1 + \lambda)^2 T_c^2$, where γ_{band} is an unrenormalized band value.²⁷⁻²⁹⁾ Using $(1 + \lambda) = 4.2$ and 7.6 determined from the dHvA measurements and $\gamma_{band} = 11.1$ and 9.6 $\text{mJK}^{-2}\text{mol}^{-1}$ for CsOs_2O_6 and KOs_2O_6 ,^{5,18)} respectively, we can estimate the ratio of $B_{c2}^*(0)$ between the two compounds to be 21. The agreement between the observed and estimated factors indicates that $B_{c2}(0)$ is basically determined by orbital effects in these compounds with only minor influence of spin effects, as previously proposed in refs. 7 and 30. Numerical estimations can be made from the formula $B_{c2}^*(0) = -0.73 dB_{c2}/dT|_{T=T_c} T_c$.²⁷⁾ Using experimental values of $-dB_{c2}/dT|_{T=T_c} = 0.44$ and 3.61 T/K,^{5,14,30)} we have $B_{c2}^*(0) = 1.0$ and 25 T for CsOs_2O_6 and KOs_2O_6 , respectively. The fact that $B_{c2}(0) > B_{c2}^*(0)$ may be accounted for by multi band (KOs_2O_6 is a two-band system) and Fermi surface effects as proposed in ref. 7.

Next we consider how the large $B_{c2}(0)$ can be reconciled with the small B_{po} . When electron-phonon and electron-electron interactions exist, the paramagnetic critical field B_p is modified from the BCS one B_{po} : $B_p = B_{po}(1 + \lambda)/S$, where S is the Stoner enhancement factor of the spin susceptibility.^{29,31)}

In a previous work,¹⁸⁾ we have determined the product Sg , where g is the g factor, to be 9.4 for the β orbit in CsOs_2O_6 . If we assume $g = 2$, we have $S = 4.7$. Using this value and $(1 + \lambda) = 7.6$, we obtain $B_p = 29$ T for KOs_2O_6 , which is still insufficient to explain the observed $B_{c2}(0)$.

In this argument, S may have been overestimated, because, in general, electrons circuiting a small orbit that crosses the Brillouin zone boundary several times as the present β orbit may have a g value much larger than 2.²¹⁾

We therefore switch to values of S determined from bulk magnetic susceptibility measurements.^{5,14)} $S = 3.1$ for CsOs_2O_6 in ref. 18 whereas $S = 1.3$ and 1.2 for CsOs_2O_6 and KOs_2O_6 , respectively, in ref. 5. In the former, a possible contribution of the Van Vleck term was neglected, while in the latter the Van Vleck term was assumed to be the same as that in $\text{Cd}_2\text{Re}_2\text{O}_7$, which was determined from the K (Knight shift)- χ plot in Cd NMR measurements,³²⁾ and was subtracted from the measured total susceptibilities. With $S = 3.1$ or 1.2, we have $B_p = 43$ or 112 T, respectively.

In order to see a combined effect of $B_{c2}^*(0)$ and B_p , we may resort to a dirty-limit formula, $B_{c2}(0)^{-2} = B_{c2}^*(0)^{-2} + 2B_p^{-2}$.^{29,31)} If we assume $B_{c2}^*(0) = 33$ T (rather arbitrarily), we have $B_{c2}(0) = 22$ or 30 T for $B_p = 43$ or 112 T, respectively. Although impurity spin-orbit scattering may increase thus calculated $B_{c2}(0)$ values, such an effect should be small for the present high-quality crystal. It is clear that $B_p = 112$ T that was derived with $S = 1.2$ is more appropriate. We therefore conclude that, to be consistent with the observed large $B_{c2}(0)$, the Stoner enhancement factor must be close to 1 in KOs_2O_6 as estimated in ref. 5. This immediately implies that the observed mass enhancement $(1 + \lambda) = 7.6$ is due to electron-phonon interactions, including electron-rattling-mode ones, for the most part: Let us assume $(1 + \lambda) = (1 + \lambda_{ep})(1 + \lambda_{ee})$, where λ_{ep} and λ_{ee} are an electron-phonon and an electron-electron coupling constant, respectively. Effects of electron-rattling-mode interactions are included in the former. In general, $S/(1 + \lambda_{ee}) > 1$.³³⁾ Therefore, if $S = 1.2$, $(1 + \lambda_{ee}) < 1.2$, and hence $(1 + \lambda_{ep}) > 6.3$.

In conclusion, we have performed magnetic torque measurements on KOs_2O_6 . We have confirmed that the upper critical field amounts to ~ 30 T. We have observed de Haas-van Alphen oscillations arising from the β orbit on the hole Fermi surface. The size and angular dependence of the β orbit are in excellent agreement with the band structure calculation. A large mass enhancement of $(1 + \lambda) = m^*/m_{band} = 7.6$ has been found. Considering how the large B_{c2} can be reconciled with Pauli limiting, we have concluded that the Stoner enhancement factor must be close to one and hence that the observed mass enhancement must be of electron-phonon origin, including electron-rattling-mode interactions, for the most part.

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